EUROPEAN PLASMA RESEARCH ACCELERATOR WITH EXCELLENCE IN APPLICATIONS



EuPRAXIA@SPARC_LAB machine upgrade & additional beamlines

Francesco Stellato – Università Tor Vergata & INFN On behalf of EuPRAXIA@SPARC_LAB WA8

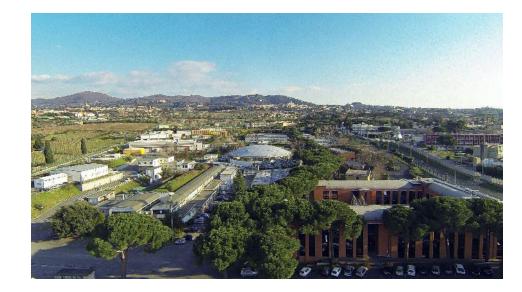




This project has received funding from the European Union's Horizon Europe research and innovation programme under grant agreement No. 101079773

Outline

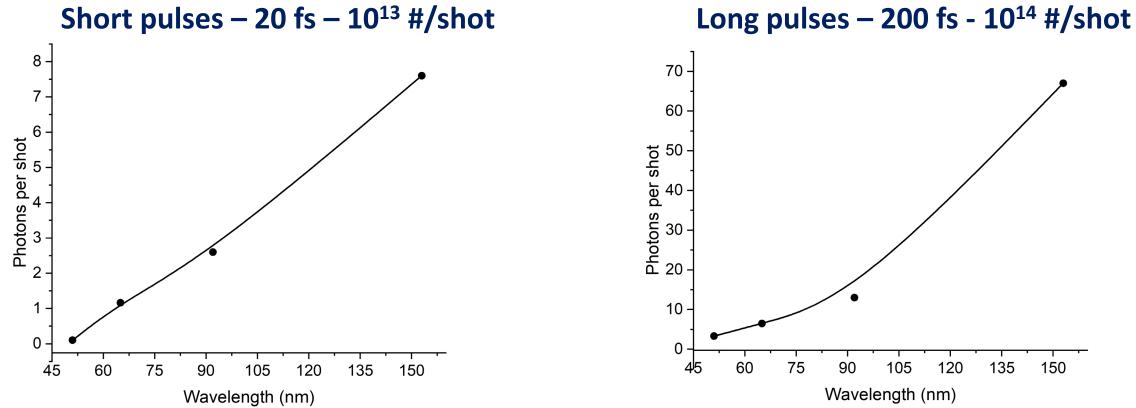
- What is ARIA?
 - Wavelength, brilliance & Co.
- What are the similar sources around the world? Synchrotron, FEL and HHG sources
- What is the photon science that can benefit from ARIA? Experimental techniques & sample classes
- What will ARIA look like?
 - Overview of the experimental endstation



ARIA is a VUV-seeded FEL beamline that will be part of the EuPRAXIA@SPARC_LAB user facility

ARIA will deliver ultra-bright, ultra-short photon pulses in the 50 to 180 nm wavelength range (7-25 eV), with tunable linear and circular polarization.

Parameter	Value
Wavelength	50-180 nm
Photons/pulse	10 ¹³ - 10 ¹⁴
Pulse duration	20/200 fs
Repetition rate	100 Hz

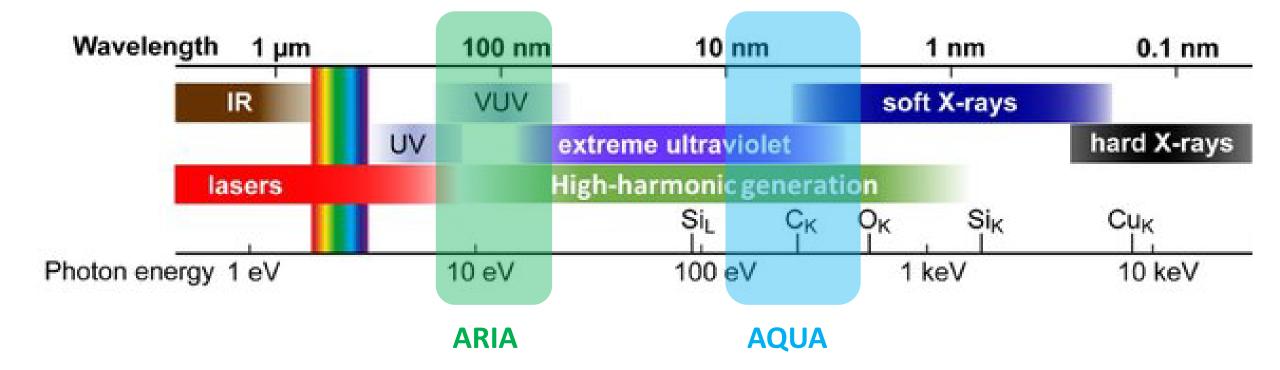


Courtesy of M. Opromolla

ARIA will be complementary to the other EuPRAXIA@SPARC_LAB FEL beamline, **AQUA**

AQUA is a soft-X-ray SASE FEL beamline that will deliver photons in the region between 4 and 10 nm

Parameter	Value
Wavelength	4-10 nm
Photons/pulse	10 ¹⁰ - 10 ¹¹
Pulse duration	< 50 fs
Repetition rate	100 Hz

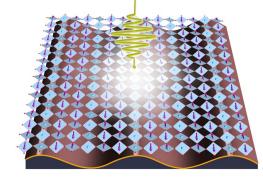


Science @ AQUA in a nutshell

Coherent imaging in the water window, including stereoimaging schemes

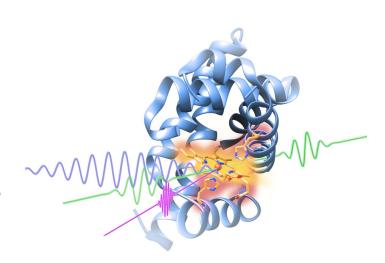
Hydrated environment measurements on bacteria, viruses, nanoparticles, ashes...

X-ray & Photoemission pump-probe spectroscopy



Ultrafast studies on hydrocarbons, aminoacids, alloys, warm-dense matter, cuprates, catalysts, batteries

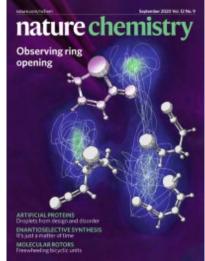
Ultrafast **Raman spectroscopy** on metalloproteins and organometallic molecules



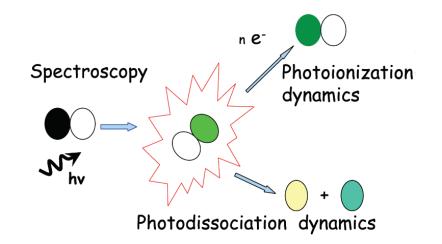
Science @ ARIA in a nutshell

Pump-probe studies of molecules photofragmentation



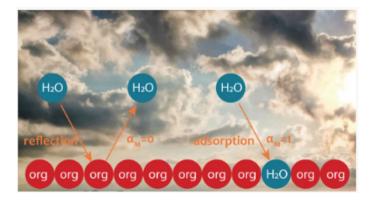


VUV, ion & Photoemission spectroscopy

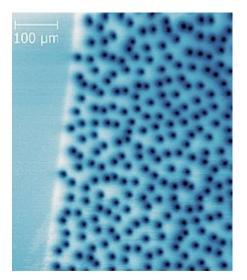


Studies on molecules in gas phase, including astrochemistry

Aerosol, Liquids and solution chemistry



Solids phase: superconductors and magnetic materials, hydrodynamics of lattices.

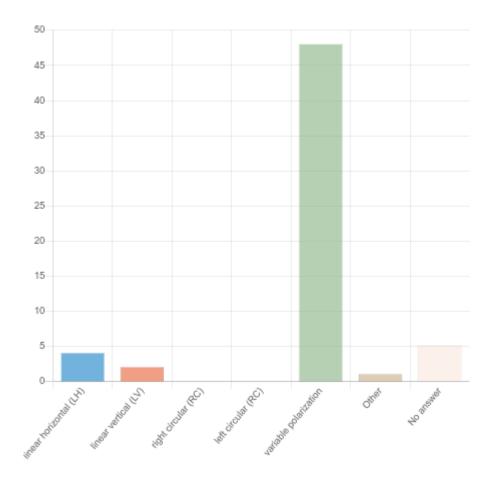


Requests from the EuPRAXIA-PP users' survey

Photon wavelength is a crucial parameter:

- 33% prefer EUV (< 300 eV)
- 47% prefer soft x-rays
- 20% prefer hard x-rays (> 7 keV)

The long-wavelength tail of the EUV falls within the **ARIA** region



VUV sources around the world

Key Source Characteristics High Harmonic Generation sources:

•Photon Flux: Typically between 10⁶–10¹⁰ photons/s, lower than synchrotrons and FELs but with high coherence. •Energy Tunability: Limited tunability, with discrete harmonics based on the driving laser frequency.

•Pulse Length: Attosecond to femtosecond pulses, significantly shorter than synchrotrons and comparable to FELs. •Repetition Rate: High repetition rate, from kHz to MHz

•Compactness: Tabletop-sized, making it more accessible for laboratory use.

These sources are ideal for ultrafast time-resolved spectroscopy, attosecond science, and nanoscale imaging in the VUV range.

- Ultrashort Pulses: Attosecond to femtosecond pulses are ideal for capturing electron dynamics.
- **Coherent Light**: High spatial and temporal coherence for techniques like coherent diffractive imaging (CDI).
- **Compact & Accessible**: Tabletop-sized, making them accessible in small-scale labs and university settings.
- Attosecond Science: Unique capability for studying electron dynamics on attosecond timescales, outperforming synchrotrons/FELs in this regard.
- Lower Photon Flux: Significantly lower photon flux (10⁶–10¹⁰ photons/s), limiting use in applications requiring high signal strength.
- Limited Tunability: Tuning is more discrete (based on harmonic orders), which may limit element-specific or resonant studies.
- **Specialized Applications**: While excellent for ultrafast and coherent imaging, HHG sources may not be suitable for broader applications that require high photon flux and tunability, such as X-ray scattering or high-resolution X-ray absorption.

Key Source Characteristics Large Scale Facilities (Synchrotrons & FELs):

Photon Flux: Typically between 10¹²-10¹⁴ photons/s (synchrotrons) and 10¹⁴-10¹⁶ photons/pulse (FELs).
Energy Tunability: Wide tunability in both VUV and soft X-ray regions, allowing for resonant and element-specific studies.
Pulse Length: Synchrotrons offer ps-range pulses, while FELs provide fs-range pulses, ideal for ultrafast time-resolved experiments.

These facilities are crucial for conducting high-resolution, high-flux, and tunable experiments in the VUV range, often required for advanced material science, surface science, molecular dynamics, and time-resolved studies.



- High Photon Flux: Orders of magnitude higher photon flux (10¹²-10¹⁶ photons/s or per pulse), suitable for high-sensitivity and high-resolution experiments.
- Wide Energy Tunability: Allows for resonant, element-specific studies across a broad energy range.
- Femtosecond Pulse Duration: FELs offer ultrafast pulses in the fs range for time-resolved experiments.
- Established Infrastructure: Access to advanced instruments for diverse techniques like spectroscopy, scattering, and imaging.
- Limited Accessibility: Large, centralized facilities often require competitive proposals for beam time, limiting frequent use.
- Size & Complexity: Large-scale, requiring significant infrastructure and operation costs.
- **Pulse Duration Limitation**: Synchrotrons are limited to the tens of ps timescale, FELs can reach the fs timescale, but (so far) cannot reach the attosecond timescales achievable by HHG sources.

Synchrotron beamlines in the VUV

Some examples

DE BESSY II - Helmholtz-Zentrum Berlin (Germany)

Source Type: Synchrotron Radiation Facility **Photon Energy Range:** 2 eV to 10 keV (VUV to soft X-rays) **Photon Flux:** 10¹³ – 10¹⁵ photons/s

Energy Tunability: Wide range tunability, optimized for VUV and soft X-rays **Pulse Length:** 50 ps

Specific Beamlines:

- **UE52-PGM Beamline:** Focuses on VUV and soft X-ray absorption spectroscopy.
- UE112 Beamline: For high-resolution PES and ARPES in the VUV energy range.

Applications: Photoemission, ARPES, surface science, catalysis research, material science.

Samples: Solids (surfaces and thin films), liquids (under certain conditions), and gas phase (with appropriate sample preparation).

SE MAX IV Laboratory (Sweden)

Source Type: Synchrotron Radiation Facility Photon Energy Range: 10 eV to 40 keV (VUV to hard X-rays)

Photon Flux: 10¹³ – 10¹⁴ photons/s

Energy Tunability: Fully tunable across the VUV range

Pulse Length: Sub-100 fs in single-bunch mode, or 100 ps in multi-bunch mode **Specific Beamlines:**

 FinEstBeAMS Beamline: Specializes in VUV and soft X-ray studies of gas-phase molecules and surfaces.

Applications: VUV absorption spectroscopy, PES, time-resolved experiments, molecular dynamics, Rydberg state studies.

Samples: Solids (surfaces, thin films, and bulk), liquids (often in combination with in situ cells), and gas phase.





US Advanced Light Source (ALS) - Lawrence Berkeley National

Laboratory (USA)

Source Type: Synchrotron Radiation Facility **Photon Energy Range:** 10 eV to 2000 eV (covers VUV to soft X-rays) **Photon Flux:** 10¹² – 10¹⁴ photons/s

Energy Tunability: Tunable over a wide range using various beamlines **Pulse Length:** 70 ps

Specific Beamlines:

- Beamline 6.3.2 specializes in VUV absorption and surface science.
- High-resolution PES and ARPES are available at multiple beamlines.

Applications: PES, ARPES, VUV spectroscopy, surface science, material characterization.

Samples: Solids (surfaces, thin films), liquids (under special conditions), and gas phase.



FR Synchrotron SOLEIL (France)

Source Type: Synchrotron Radiation Facility

Photon Energy Range: 5 eV to 40 keV (covers VUV, soft X-rays, and hard X-rays) **Photon Flux:** $10^{13} - 10^{15}$ photons/s

Energy Tunability: Full energy tunability with undulators and monochromators **Pulse Length:** 50–100 ps

Specific Beamlines:

- **DESIRS Beamline:** Dedicated to VUV spectroscopy (5–40 eV), providing high photon flux and energy resolution.
- **PLEIADE PES Beamline:** Offers angle-resolved and resonant PES experiments.

• **DISCO Beamline:** Offers angle-resolved and resonant PES experiments. **Applications:** VUV photoemission, ARPES, molecular physics, astrophysics, surface chemistry.

Samples: Solids (surfaces, thin films, bulk), liquids (special setups), and gas phase.



CA Canadian Light Source (CLS) - VUV Beamlines (Canada)

Source Type: Synchrotron Radiation Facility **Photon Energy Range:** 10 eV to 2500 eV (VUV to soft X-rays) **Photon Flux:** 10¹³ – 10¹⁴ photons/s

Energy Tunability: Wide energy tunability with various beamlines

Pulse Length: 80 ps

Specific Beamlines:

• VLS-PGM Beamline: Specialized in high-resolution VUV spectroscopy and PES studies.

Applications: VUV absorption, PES, surface science, biomolecular structure studies.

Samples: Solids (surfaces, thin films, and bulk), liquids (with appropriate setups), and gas phase.

CH PSI - Swiss Light Source (Switzerland)

Source Type: Synchrotron Radiation Facility **Photon Energy Range**: 5 eV to 1500 eV (VUV to soft X-rays)

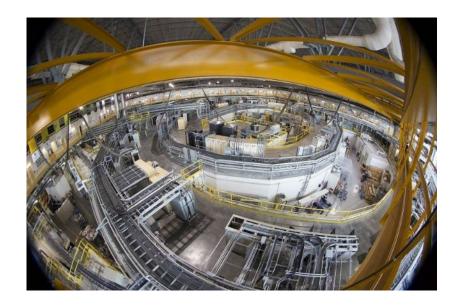
Photon Flux: $10^{12} - 10^{14}$ photons/s

Energy Tunability: Tunable over a wide range using insertion devices (undulators) **Pulse Length**: 80 ps

Specific Beamlines:

X07MA and X04DB Beamlines : Specialize in VUV and soft X-ray spectroscopy, with a focus on spectromicroscopy and imaging.
 Applications: VUV absorption, PES, surface science, biological studies, nanomaterials.

Samples: Solids (surfaces, thin films, and bulk), biological materials, and gas phase.





Elettra Sincrotrone Trieste (Italy)

Source Type: Synchrotron Radiation Facility **Photon Energy Range:** 8 eV to 15 keV (VUV to soft and hard X-rays)

Photon Flux: $10^{13} - 10^{15}$ photons/s

Energy Tunability: Tunable over a wide range using insertion devices (undulators, wigglers)

Pulse Length: 100 ps

Specific Beamlines:

- **APE Beamline:** Focuses on VUV and soft X-ray ARPES and PES studies.
- **SuperESCA Beamline:** Dedicated to high-resolution PES and surface science.
- MOST@Elettra 2.0 (GasPhase +Circular Polarization)

Applications: PES, ARPES, VUV absorption, surface chemistry, material science, thin-film studies.

Samples: Solids (surfaces, thin films, and bulk), liquids (in specialized environments), and gas phase.



FEL beamlines in the VUV

Some examples

FERMI Free-Electron Laser (Italy)

Source Type: Free-Electron Laser (FEL) **Photon Energy Range:** 12 eV to 310 eV (covers VUV and soft X-

rays)

Photon Flux: 10¹¹ – 10¹⁴ photons/pulse

Energy Tunability: Tunable across a wide energy range using FEL amplification

Pulse Length: Sub-100 fs (femtoseconds), ideal for time-resolved studies

Specific Beamlines:

- **EIS-TIMEX Beamline:** For time-resolved spectroscopy and coherent VUV studies.
- **LDM Beamline:** For laser-driven dynamic experiments in the VUV range.





Applications: Time-resolved spectroscopy, ultrafast dynamics, pump-probe experiments, photoionization studies, VUV spectroscopy.

Dalian Coherent Light Source (DCLS) (China)

SourceType:Free-ElectronLaser(FEL)Photon Energy Range:8 eV to 24 eV (covers VUV range)Photon Flux: $10^{11} - 10^{13}$ photons/pulseEnergy Tunability:Tunable across a limited VUV energyrange, particularly for molecular and material studiesPulseLength:Sub-femtosecond, ideal for ultrafastdynamicsandspecific Beamlines:

•**VUV Beamline:** Primarily used for studying valence electronic structures, surface chemistry, and molecular dynamics.





Applications: Time-resolved spectroscopy, photoemission, surface science, ultrafast dynamics, and VUV spectroscopy.
Samples: Solids (surfaces, thin films), liquids (with special sample environments), and gas phase.

FLASH Free-Electron Laser (Germany)

Source Type: Free-Electron Laser (FEL) Photon Energy Range: 10 eV to 250 eV (VUV and soft X-rays) Photon Flux: 10¹¹–10¹⁴ photons/pulse Energy Tunability: Wide tunability across the VUV and soft Xray regions Pulse Length: Sub-100 fs (femtoseconds), ideal for time-

resolved experiments

Specific Beamlines:

FLASH1 and FLASH2 Beamlines: Offer VUV and soft X-ray light for pump-probe and time-resolved spectroscopy experiments.





Applications: Ultrafast dynamics, photoemission spectroscopy, molecular dissociation, surface studies, Rydberg state research.
Samples: Gas phase (often used for isolated molecules), solids (with advanced techniques), and liquids (under specific conditions).

Science in the **ARIA** range

Atomic, Molecular, and Cluster Physics:

Experiments on atomic, molecular, and cluster systems, covering gas adsorbates, interfaces, and the evolution of properties from isolated particles to condensed phases.

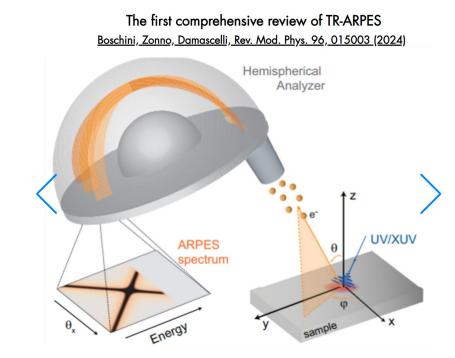
Photo-Ionization and Atmospheric Science:

Access to ionization thresholds and ionic states allows the study of atmospheric species (from the troposphere to ionosphere) and combustion species. **Electronic Structure and Excited States is solids**: Electronic transitions in materials like nano-carbons, metal oxides. Excitons, polarons \rightarrow spintronics & solar cells.

Chirality and Molecular Dynamics:

Exploring chirality and natural dichroism in biological systems

ARIA : 7-25 eV



Techniques

- Photoemission Spectroscopy
- (also Angle-Resolved)
- Time-of-Flight ion spectroscopy (ToF) / Velocity Map Imaging (VMI)
- ...

Atomic, Molecular, and Cluster Physics

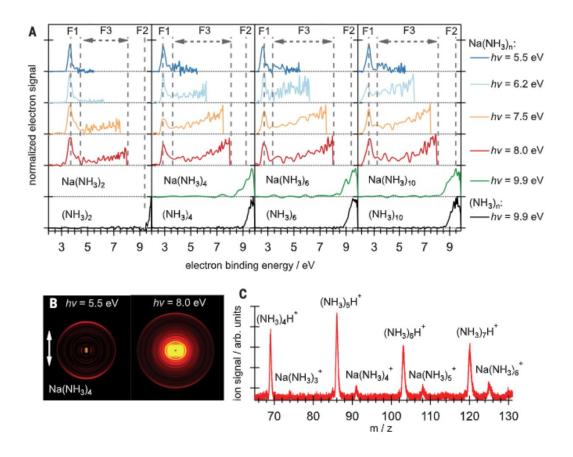
ARIA will enable advanced experiments on **atomic, molecular, and cluster** systems, covering gas adsorbates, interfaces, and the evolution of properties from isolated particles to condensed phases. Studies range from rare gases to elements with high vaporization temperatures.

[1] Hartweg, Sebastian, et al. "Solvated dielectrons from optical excitation: An effective source of low-energy electrons." *Science* 380.6650 (2023): 1161-1165.

[2] Daniely, Amit, et al. "A Vacuum Ultraviolet Photoionization Mass Spectrometry and Density Functional Calculation Study of Formic Acid–Water Clusters." *The Journal of Physical Chemistry A* 128.31 (2024): 6392-6401.

[3] Rupp, Daniela, et al. "Recombination-enhanced surface expansion of clusters in intense soft x-ray laser pulses." *Physical review letters* 117.15 (2016): 153401.

ARIA : 7-25 eV

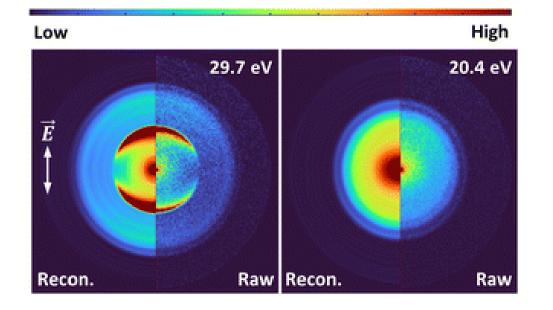


Low-energy electrons dissolved in liquid ammonia or aqueous media promote challenging reduction reactions, but can also cause radiation damage to biological tissue. **Ultraviolet photoexcitation** (5.5 – 10 eV) of metal-ammonia clusters could be used to generate tunable low-energy electrons in situ. [1]

Molecules in the gas phase

ARIA will allow for the study of **gas phase molecules** to probe electronic transitions and gain insights into molecular structure and dynamics. This facilitates the investigation of ionization processes and fragmentation pathways, providing a deeper understanding of chemical reactions in the gas phase

ARIA : 7-25 eV



Oxybenzone in the gas phase can be characterized by mass spectrometry and angle-resolved photoelectron spectroscopy, using both single and multiphoton ionization schemes.

A tabletop high harmonic generation source with a monochromator was used for single-photon ionization of oxybenzone with photon energies of up to 35.7 eV.

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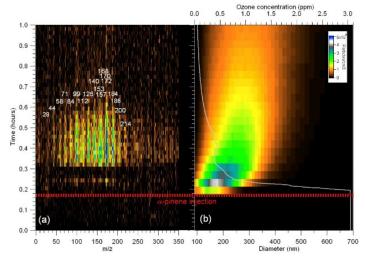
Tsizin, S., Ban, L., Chasovskikh, E., Yoder, B. L., & Signorell, R. (2024). Valence photoelectron imaging of molecular oxybenzone. *Physical Chemistry Chemical Physics*, *26*(28), 19236-

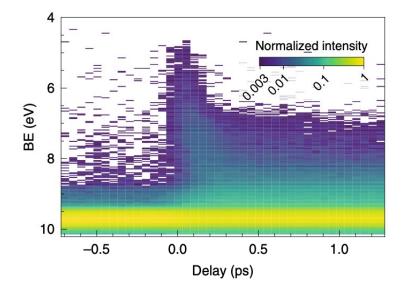
Photo-Ionization and Atmospheric Science:

Access to **ionization thresholds** and ionic states allows for the study of **atmospheric species** (from the troposphere to ionosphere), poorly understood **combustion species**, and biological materials. Techniques like **mass spectrometry** and **photo-emission** are employed. Studies range from rare gases to elements with high vaporization temperatures, gas adsorbates, interfaces, and the evolution of properties from isolated particles to condensed phases,

A reaction chamber coupled with a **photoionization aerosol time-of-flight mass spectrometer** was used for realtime analysis of **organic and mixed aerosols**, leveraging synchrotron vacuum ultraviolet photons (8 eV) for ionization. The 3D mass spectrum over time is obtained by sampling an aerosol [1]

ARIA : 7-25 eV





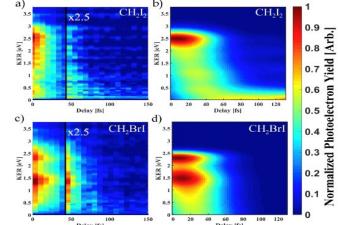
Time-resolved photoelectron spectroscopy with a seeded extreme ultraviolet free-electron laser (19.2 eV) to trace the ultrafast ring opening of gas-phase thiophenone molecules following ultraviolet photoexcitation. [2]

 Baeza-Romero, María Teresa, et al. "A smog chamber study coupling a photoionization aerosol electron/ion spectrometer to VUV synchrotron radiation: organic and inorganic-organic mixed aerosol analysis." *The European Physical Journal D* 70 (2016): 1-11.
 Pathak, Shashank, et al. "Tracking the ultraviolet-induced photochemistry of thiophenone during and after ultrafast ring opening." *Nature chemistry* 12.9 (2020): 795-800.
 Mason, N. J., et al. "Atmospheric chemistry with synchrotron radiation." *Journal of Physics B: Atomic, Molecular and Optical Physics* 38.9 (2005): S893.

Electronic Structure and Excited States:

Resonant techniques in the VUV range provide insights into electronic transitions in materials like nano-carbons and metal oxides. ARIA would also support **two-photon photoemission** (2PPE) for studying **excitons**, **polarons**, and states relevant to spintronics batteries and solar cells.

Excited state dynamics of **diiodomethane and bromoiodomethane** are studied using **time resolved PES**. A 4.65 eV UV pump pulse excites both molecules and the dynamics are probed via photoionization using a 7.75 eV probe pulse. [1]

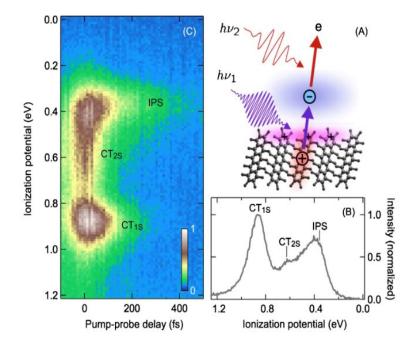


[1] Horton, Spencer L., et al. "Excited state dynamics of CH2I2 and CH2BrI studied with UV pump VUV probe photoelectron spectroscopy." *The Journal of chemical physics* 150.17 (2019).

[2] Chen, Jiao-Jiao, et al. "A VUV photoionization time-of-flight mass spectrometer for the formation, distribution, and reaction of nano-sized neutral metal oxide clusters." *International Journal of Mass Spectrometry* 422 (2017): 98-104.

[3] Zhu, X-Y. "Photoemission from excitons in organic semiconductors." *Journal of Electron Spectroscopy and Related Phenomena* 204 (2015): 75-79.





PES can be a powerful tool in providing a unique view on the energetics and dynamics of excitons in semiconductor materials. The experiment relies on **timeresolved two-photon photoemission** (TR-2PPE) spectroscopy in which the first laser pulse creates excitons in a **semiconduttive material** and the second laser pulse ionizes the excitons for detection. The pump is in the range of 2.15 - 4.65 eV [3]

Electron dynamics in solids

Angle-Resolved Photo Emission Spectroscopy (ARPES) experiments can provide detailed information about the electronic dynamics and the underlying properties of materials.

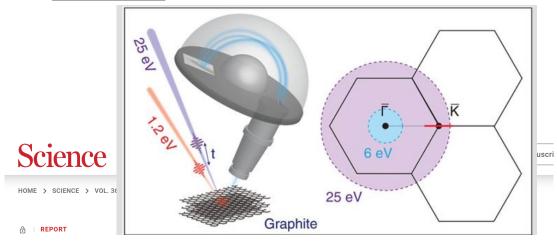
Exploration of ultrafast electronic dynamics, orbital symmetries, surface vs. bulk states, and spin-dependent phenomena. ARIA will enable the access to **multiphoton processes** and **non-linear optics**.

ARIA : 7-25 eV



Journal of Electron Spectroscopy and Related Phenomena Volume 243, August 2020, 146978

Time-resolved VUV ARPES at 10.8 eV photon energy and MHz repetition rate



Direct determination of mode-projected electronphonon coupling in the time domain

M.X. NA (D, A.K. MILLS (D, F. BOSCHINI (D, M. MICHIARDI (D, B. NOSARZEWSKI (D, R. P. DAY (D, E. RAZZOLI (D, A. SHEYERMAN (D, M. SCHNEIDER (D, [...], AND A. DAMASCELLI (D +5 authors) Authors Info & Affiliations

Time- and angle-resolved photoemission spectroscopy (TR-ARPES) is applied to **graphite**, focusing on the dynamics of photoinjected electrons at the K point.

Chirality and Molecular Dynamics:

ARIA's capabilities will extend to exploring chirality and natural dichroism in biological systems, aiding in the understanding of molecular structures.

The valence shell photoionization of the **simplest chiral amino acid**, **alanine**, is investigated over the vacuum ultraviolet region from its ionization threshold. Tunable and variable polarization synchrotron radiation was coupled to a **double imaging photoelectron/photoion coincidence** (i 2 PEPICO) spectrometer to produce mass-selected threshold photoelectron spectra and derive thestate-selected fragmentation channels. Photon energy is in the range in 9-15 eV [2].

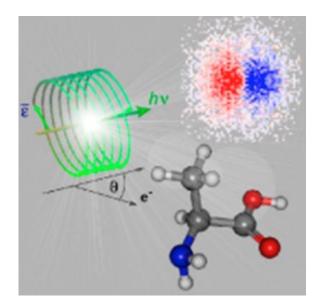
[1]Nahon, Laurent, et al. "Determination of accurate electron chiral asymmetries in fenchone and camphor in the VUV range: sensitivity to isomerism and enantiomeric purity." *Physical Chemistry Chemical Physics* 18.18 (2016): 12696-12706.

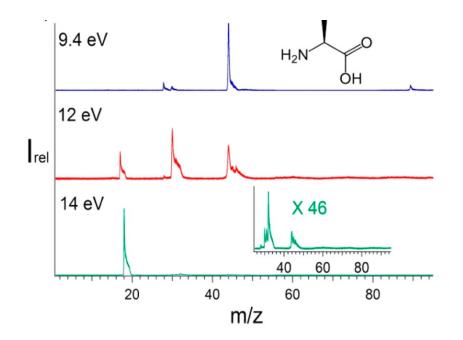
[2] Tia, Maurice, et al. "VUV photodynamics and chiral asymmetry in the photoionization of gas phase alanine enantiomers." *The Journal of Physical Chemistry A* 118.15 (2014): 2765-2779.

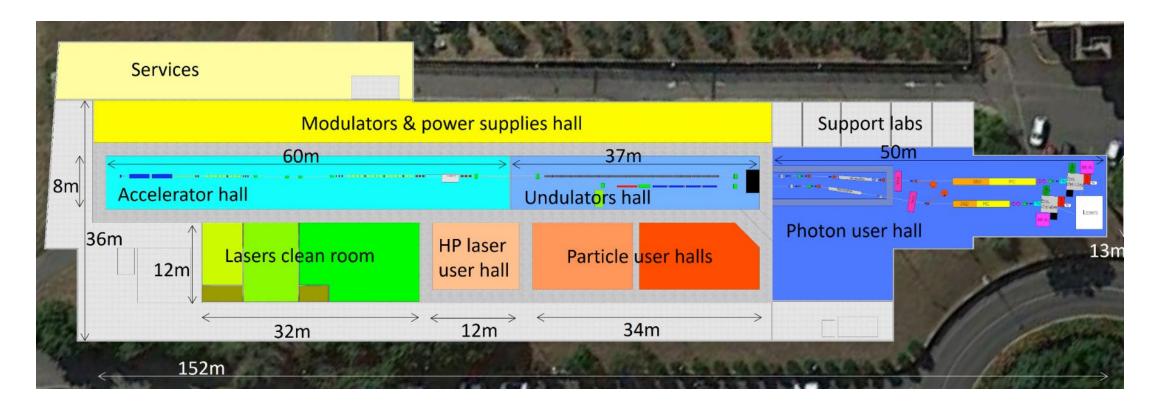
[3] Mason, N. J., et al. "Atmospheric chemistry with synchrotron radiation." *Journal of Physics B: Atomic, Molecular and Optical Physics* 38.9 (2005): S893.

[4] Nahon, Laurent, et al. "DESIRS: a state-of-the-art VUV beamline featuring high resolution and variable polarization for spectroscopy and dichroism at SOLEIL." *Journal of synchrotron radiation* 19.4 (2012): 508-520.



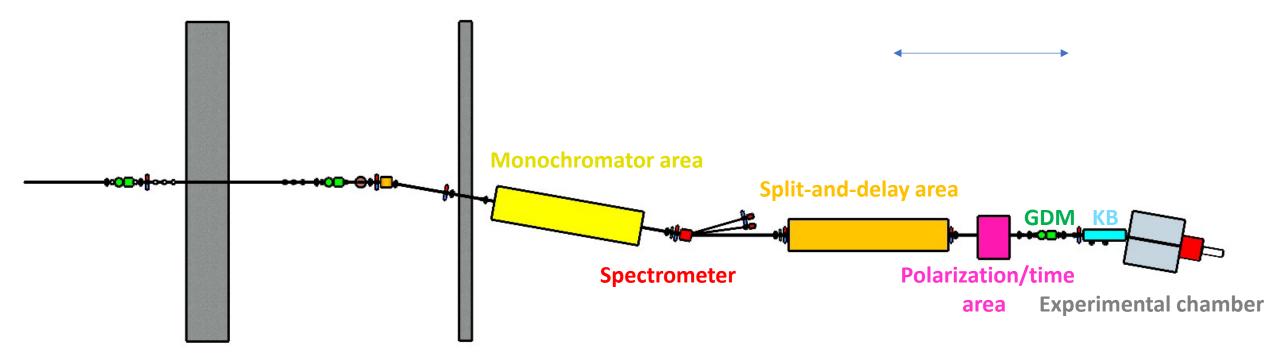






Layout of the EuPRAXIA@SPARC_LAB facility

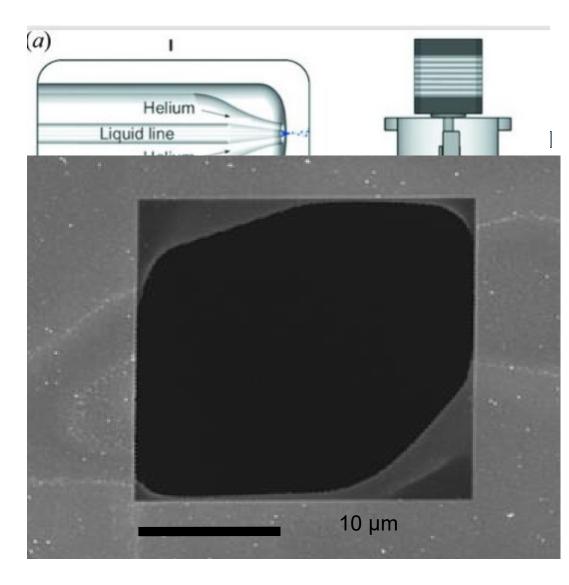
After the undulator, approximately 50 m of the beamline will deliver the photon beam to an experimental chamber. All of its elements can be divided into two different conceptual groups: beam transport and beam diagnostics.



Courtesy of F. Villa

Sample delivery systems (to be shared with AQUA)

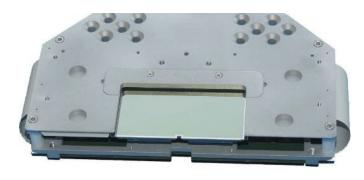
- Liquid jets (micro-jets, based on the gas dynamic virtual nozzle technology, including those developed for sample mixing and for a lower sample consumption at pulsed sources)
- Aerosols sources for gas-phase measurements
- Solid samples mounted on motorized micro-precision stages and/or hexapod-like devices. Grids for single-shot expose-and-destroy measurements.



Detectors (some to be shared with AQUA)

Photons

- 2D, small pixel, fast-readout, high dynamic range detector



 OD & 2D energy-resolved detector for spectroscopy

Electron & Ions

High position sensitivity, large detection area, fine gridding to allow reconstruction of threedimensional arrangement and motion of atoms within a molecule and distinguish between ions of different masses and charge states

An inlet for an **external laser** will allow for the performing of laser pump/FEL– probe time-resolved experiments.



Two external **laser sources** (to be shared with **AQUA**) are foreseen – but **open for discussion**!

- 1- **Ti:sapphire** fixed wavelength (800nm) femtosecond laser, with also the possibility of exploiting second (400 nm) and third (267nm) harmonics.
- 2- Optical Parametric Amplifier (OPA), with tensof femtosecond pulses in the 700-900 nm range.Options to go from UV to MIR.

For this starting point, thanks to

(for discussions, ideas and inputs for this presentation)

Marcello Coreno Emiliano De Santis Luca Giannessi Augusto Marcelli Michele Opromolla Emiliano Principi Fabio Villa All people in WA8

You all for the kind attention